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CHEMICAL AND ISOTOPIC COMPOSITIONS IN ACID
RESIDUES FROM VARIOUS METEORITESN. Kano¹, K. Yamakoshi¹, H. Matsuzaki¹ and K. Nogami²

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To study the pre-history of the solar system such as possible direct evidences for processes of nucleosynthesis in the pre-solar stage and detection of extinct radioactive nuclide, we are planning to carry out systematic isotopic investigations on Ru, Mg and so on in primordial samples. In this paper, we present the results of chemical compositions of acid residues obtained from three types of meteorites [1) Canyon Diablo (IA), 2) Allende (CV3), 3) Nuevo Mercuro (H5)] and the preliminary results of Ru isotopic compositions.

Many studies on isotopic anomalies of elements, specially noble gases, in acid residues from primitive meteorites have been done [e.g. 1-3]. Particularly, Zinner et al. have recently recovered silicon carbide from therein, and detected isotopic anomalies of noble gases, C, N, Si, Mg, Ba etc. [4-7]. Therefore, these samples are considered to be of precursor origins which have survived the pre-solar nebula stages.

The method to recover acid residues from meteorites in our work are as follows; After removing fusion crusts with a ceramics blade grinder, the three types of meteorites were disaggregated by using the freeze-thaw method and were dissolved by repeated alternating treatments with HCl, HF, and aqua regia at room temperature. The acid residues were concentrated by centrifuging and then dried up. The weight of the acid residues we obtained are listed in Table 1 with the total weight of the bulk meteorite.

Elemental analyses were carried out by combustion method, INAA and AAS. The typical results of elemental analyses are shown in Table 2 and Fig. 1. Fig. 1 show that refractory elements are generally enriched in acid residues regardless of the kind of meteorites. It matches the features that acid residues contain primary condensates from the cooling solar gas as well as extra-solar grains. Particularly, in acid residue of Allende, enrichment factors for refractory siderophiles are generally higher (7-20 times/CI), however in the cases of W and Mo, they are slightly depleted compared with those of Canyon Diablo. Since both elements would be the first metals to be oxidized under high oxygen fugacity [e.g. 8,9], acid residue of Allende should contain fractions that were produced under oxidising conditions. And the considerable diverse chemical compositions of acid residues between these meteorites deserves our attention. It may arise from the difference of generative environmental conditions of these meteorites in space.

The samples were decomposed in sealed teflon vessels by a microwave dissolution method and then Ru was separated by distillation [10]. The total procedural Ru blank was measured by isotope dilution method using ¹⁰⁴Ru enriched spike, and only a few ng were detected. Ru isotopic analyses have been performed by a VG 354 thermal ionization mass spectrometer. Ru ion beams in each samples were so faint because of its high ionization potential (7.36eV) and small sizes of samples that they were detected by the Daly Photomultiplier detector equipped with an ion counting assembly. In mass spectrometric techniques of Ru, the zone-refined (99.995%) outgassed V-shaped Re single filament with silica gel and phosphoric acid was employed.

Fig. 2 shows preliminary results for Ru isotopic analyses. In these measurements, all ratios of Ru were corrected for mass fractionation by normalization to ⁹⁹Ru/¹⁰²Ru=0.4042 (exponential law) provisionally [11]. And Mo ion beam from Re filaments were not negligible as the filament temperature increases, so Mo isobaric interference was corrected by the monitoring of ⁹⁷Mo based on recent precise measurements of Mo isotopes [12]. In the measurement of acid residue of Allende, all ratios were not found to be distinguishable from terrestrial values within the experimental errors, though errors were rather high because of the interference of molecular ion peaks and small Ru ion beam intensities. Poths et al. [13] also reported that in two aliquants of acid residue of Allende, all isotopic ratios of Ru are indistinguishable from the terrestrial values within the experimental errors.

The fact that Ru isotopic anomalies have been not yet detected even in Allende acid residue, which is considered to be the one of the most primordial and exotic samples, suggests as follows; Ru components originate from interstellar medium might be very rare and/or the diffusion of Ru might be unexpectedly rapid due to high temperatures and shock waves or something during the condensation stage of the solar system.

However, judging from fraction % of acid residue in Allende and Fe contents (~1%) in these acid residues, it could be said that the acid treatments in this work were relatively moderate and/or incomplete. We are, therefore, going on with acid treatments still more. In other words, the acid residues we obtained here are not so definite samples. So, the chemical compositions of the acid residues differ with the kinds of acids, and the conditions

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under acid treatments.

Unfortunately, we could not identify what kind of minerals were there in these acid residues by our SEM (HORIBA EMAX-8000) this time. We are now preparing to check these acid residues by high resolution transmission electron microscopy (TEM) to identify what kind of minerals contained there. And much more precise and reliable isotopic data will be obtained, then more detailed discussions on the prehistory of the solar system, and the origin of acid residues could be done, too.

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References [1] Lewis, R. S. et al. (1983) *Science*, **222**, 1013; [2] Lugmair, G. W. et al. (1983) *Science*, **222**, 1015; [3] Loss, R. D. et al. (1990) *GCA*, **54**, 3525; [4] Zinner, E. et al. (1989) *GCA*, **53**, 3273; [5] Zinner, E. et al. (1991) *Nature*, **349**, 51; [6] Zinner, E. et al. (1991) *LPS*, **22**, 1553; [7] Amari, S. et al. (1992) *LPS*, **23**, 27; [8] Palme, H. et al. (1982) *EPSL*, **61**, 1; [9] Fegley, B. and Palme, H. (1985) *EPSL*, **72**, 311; [10] Terada, K. et al. (1975) *Bull. Chem. Soc. Jpn.*, **48**, 2567 [11] Hutcheon, I. D. et al. (1987) *GCA*, **51**, 3175; [12] Qi-Lu and Masuda, A. (1992) *J. Am. Soc. Mass Spec.*, **3**, 10; [13] Poets, H. et al. (1987) *GCA*, **51**, 1143

Table 1 The weight and fraction % of acid residues

Sample Name	Sample weight (g)	Acid Residue weight (g)	Acid Residue fraction (wt. %)
Canyon Diablo	587.50	3.6425	0.62
Allende	62.379	1.8975	3.04
Nuevo Mercurio	45.104	0.4631	1.03

Table 2 Element concentration in acid residues (%) (combustion method)

	C	N	H	S
Canyon Diablo	42.86	2.82	2.72	-----
Allende	9.70	1.80	0.51	0.60
Nuevo Mercurio	-----	0.29	-----	2.22

----- stands for below detection limit.

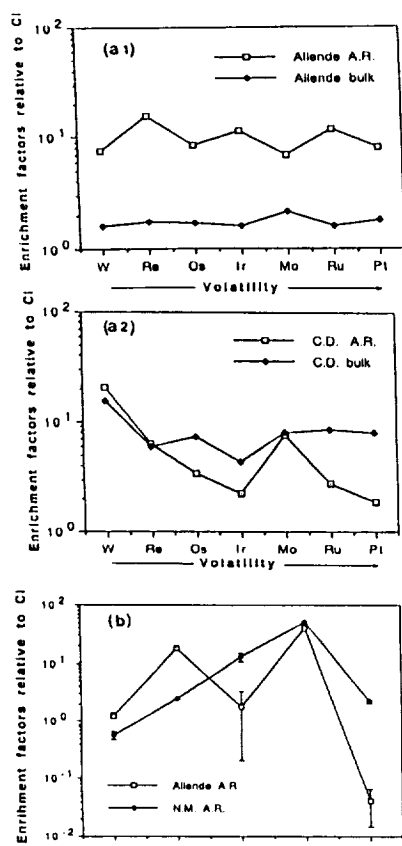


Fig. 1 Enrichment factors in acid residues relative to CI. 1 σ error bars are shown when they are larger than the symbols. A.R. stands for "Acid Residue". 3(a1), 3(a2): Refractory Siderophile 3(b): Refractory Lithophile (N.M. stands for Nuevo Mercurio)

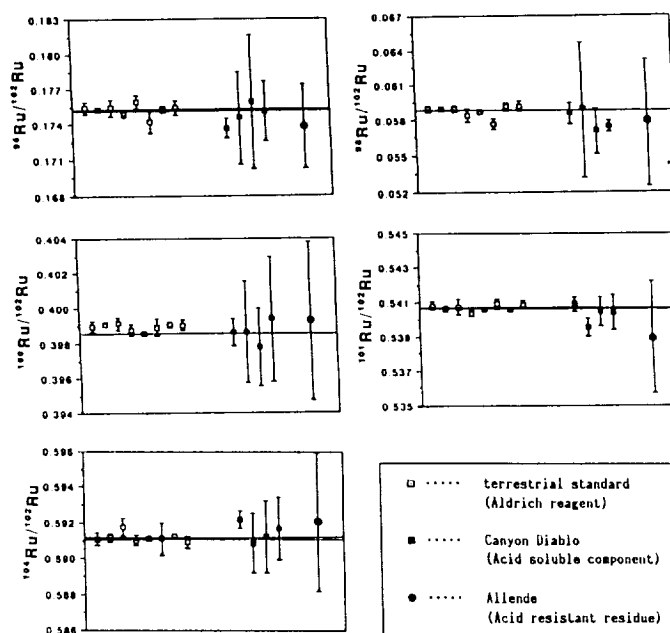


Fig. 2 Preliminary results for Ru isotopic analyses: All isotope ratios are collected for mass fractionation by normalization to $^{104}\text{Ru}/^{102}\text{Ru}=0.4042$ (exponential law). Errors are 2σ of the mean. The solid lines represent the range of weight means in terrestrial standard.